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Removal of Inorganic Trace Contaminants by Electrodialysis in A Remote Australian Community

L.J. Banasiak¹, A.I.Schäfer^{1*}

¹ School of Engineering, University of Edinburgh, Edinburgh, EH9 3JL, United Kingdom

*Tel. +44(0) 131 650 7209; Fax +44(0) 131 650 6781; email: Andrea.Schaefer@ed.ac.uk

Abstract: Water provision for developing countries is a critical issue as a vast number of lives are lost annually due to lack of access to safe drinking water. The presence and fate of inorganic trace contaminants is of particular concern. Trace inorganic contaminants have been found in elevated concentrations in drinking waters supplied directly from brackish groundwaters in developing countries. Desalination and the removal of trace inorganic contaminants from bore water sources from a remote community in Australia using electrodialysis (ED) were investigated. The influence of applied voltage on the removal of the trace contaminants was evaluated. While the results from this study demonstrated that ED is an effectual method for the removal of total dissolved solids and a number of trace inorganic contaminants from brackish groundwaters to below drinking water guideline levels, the deposition of trace contaminants on the membranes (fouling) influenced the ED process in relation to ionic flux and the effectiveness of trace contaminant removal.

Keywords: Electrodialysis, brackish groundwater, inorganic trace contaminant removal, desalination.

Introduction

Water provision in remote communities is a serious problem globally, as a large number of lives are lost annually due to lack of access to potable drinking water [1]. In Australia, approximately 67% of Indigenous communities with a population less than 100 people use bore water for their supply [2]. In these remote communities, drinking water is generally sourced from groundwater bores of varying quality, with salinity and hardness being widespread issues. Health issues related to salinity range from dehydration (as a result of reduced water consumption) to kidney dysfunction and hypertension [3, 4]. While there are no drinking water guideline values for calcium and magnesium, high levels of hardness can reduce the palatability of water and also cause water distribution pipes to block. High salt concentrations and hardness are however not the only issues facing remote communities.

The occurrence and fate of inorganic (mineral origin) trace contaminants in surface, brackish and groundwaters are also of considerable concern with regards to public health and the environment. While a number of compounds occur naturally within groundwaters, the concentration of these compounds at elevated levels and the consumption of drinking water supplied from contaminated groundwater can lead to significant health issues. The bore water investigated in this study contains a number of trace contaminants such as bromide, calcium, fluoride, magnesium, sodium, strontium and zinc. A number of these contaminants were above drinking water guidelines.

Bromide is naturally present in raw water, especially in groundwater and surface water in coastal regions, as bromide salts or as organic bromine substances [5]. Bromide is generally considered non-toxic at concentrations found in most drinking water sources; however, when water containing high levels of bromide are treated by ozonation, bromate species are formed, which are highly toxic for human health [6].

In developing countries such as Morocco, fluoride concentrations up to 20 mg/L have been found in groundwaters [7, 8], while the maximum acceptable concentration level is 1.5 mg/L [1]. The detrimental effects of excess concentrations of fluoride on teeth and the skeletal system (dental and skeletal fluorosis) is prevalent [9]. Strontium minerals are widely distributed throughout the earth and are released to the groundwater by the weathering of rocks and soils. High concentrations of strontium in groundwater of up to 53 mg/L have been reported in certain parts of Denmark. [10].

Strontium has relatively low toxicity but continued exposure may have adverse health effects. Studies have however shown that the ingestion of higher levels of strontium in drinking water prenatally may have a protective effect against caries [11]. There is also no guideline value established for strontium.

Zinc concentrations are usually low in drinking water, compared to the WHO guideline value of 3 mg/L [1]. Significant anthropogenic influences have led to increases in the bioavailability of zinc in the environment. Ingestion of enhanced zinc can cause memory impairments and copper deficiencies [12]. Elevated concentrations of zinc have also been linked to Alzheimer's disease [13].

The importance of membrane processes, such as electrodialysis (ED), for treatment of drinking water is a continuously growing field. ED has been used for a wide range of applications including desalination, the production of potable water from brackish water [14, 15], seawater [16, 17] and industrial wastewater [18, 19] and numerous other applications. The fouling of ion-exchange membranes is the main problem of electromembrane technologies. Fouling manifests itself as an increase in the electrical resistance, a decrease in the selectivity of the ion-exchange membrane and a decline in the flux of ions [20]. Scaling is the precipitation of crystalline divalent and trivalent ion hydroxides such as CaCO_3 , MgCO_3 , Mg(OH)_2 and CaSO_4 on the concentrate of the cation-exchange, and to a lesser extent on the anion-exchange membranes [21, 22]. The build-up of a scalant layer on the ion-exchange membranes leads to an increase in stack resistance, which in turn reduces the effectiveness of the ED process and leads to a decrease in membrane integrity [20].

The aim of this study is the evaluation of ED for desalination and the removal of selected inorganic trace contaminants from a brackish groundwater sourced from a remote Australian community. The focus of this study was the influence of applied voltage.

Materials and Methods

Electrodialysis System Characteristics

The ED stack used was a BEL-500 unit (Berghof, Germany) with seven Neosepta CMX-SB cation-exchange membranes (CEMs) and six Neosepta AMX-SB anion-exchange membranes (AEMs) (supplied by Eurodia, Germany; manufactured by Tokuyama Soda Ltd., Tokyo, Japan) providing for each an available membrane area of 58 cm². The ED stack was connected to a DC electric potential (GW Instek DC Power supply Model GPR-1810HD, Taiwan) through TiO_2 -coated titanium electrodes. The layout of the ED system used in the experiments is shown in Figure 1. The setup consisted of three separate solution containers for the diluate (4L), for the concentrate (4L) and for the electrode rinse (2L), which were recirculated by three peristaltic pumps.

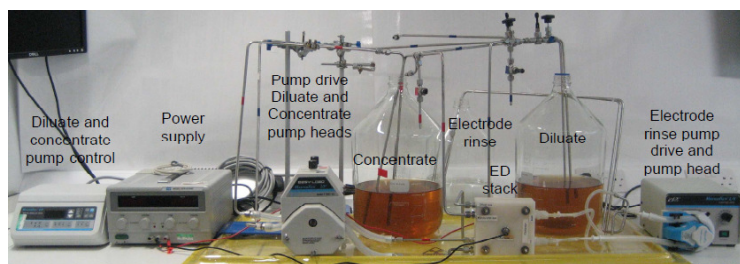


Figure 1. Electrodialysis system with pump control, power supply, peristaltic pumps, ED stack, and three containers for concentrate, diluate and electrode rinse

Bore Water Location and Characteristics

The ED experiments were performed with bore water (bore number RN13693) sourced from Pine Hill Station, a cattle farm approximately ~140 km northwest of Alice Springs, Northern Territory, Australia [23]. This bore was chosen for this study due to its poor water quality in regards to salinity, hardness and trace contaminant content. Bore water was collected, stored and transported back to the laboratory (University of Wollongong, Australia) for further use. The bore water was refrigerated before use in the ED experiments. Chemical analysis results of the bore water (undertaken at the University of Edinburgh, UK) are shown in Table 1.

Table 1. Water quality at Pine Hill Station in comparison with World Health Organization (WHO) 2006 and Australian Drinking Water Quality Guidelines (ADWG) 2004

| PARAMETER | UNIT | SAMPLE VALUE | ADWG [24] | WHO [1] |
|------------------------------|-------|---------------------|----------------------|---------|
| Bromide | mg/L | 10.6 | - | - |
| Calcium | mg/L | 105 | - | - |
| Chloride | mg/L | 2000 | 250 ^a | - |
| Fluoride | mg/L | 5.82 | 1.5 | 1.5 |
| Magnesium | mg/L | 172 | - | - |
| Potassium | mg/L | 35.4 | - | - |
| Sodium | mg/L | 1650 | 180 ^a | - |
| Strontium | mg/L | 2.28 | - | - |
| Sulphate | mg/L | 717 | 500 ^a | - |
| Uranium | mg/L | 0.261 | 0.02 | - |
| Zinc | mg/L | 0.222 | 3 ^a | - |
| TDS | mg/L | 5300 ^{b,c} | 500 ^a | - |
| pH | - | 8.4 | 6.5-8.5 ^a | - |
| Electrical conductivity (EC) | µS/cm | 8290 ^c | - | - |

^a Aesthetic guideline value. ^b Calculated using TDS = 0.064 EC [25]. ^c Average of feed conductivity throughout field testing at Pine Hill Station [26].

Experimental Protocol

To determine the influence of ED on the removal of trace contaminants from the Pine Hill bore water and the influence of applied voltage, experiments were conducted at two voltages: 12 and 18V. These voltages were chosen as a result of previous work [27] and the maximum allowable applied voltage (3 volts/cell pair). The ED system was operated in continuous operation mode during each experiment, where the feed water (initially filled in both diluate and concentrate solution tanks) was recirculated throughout the concentrate and diluate until the desired product was achieved (TDS below 500 mg/L). As a result of this, the ED experiments were not all run for the same length of time. An applied electric potential of 10V was fixed for all experiments. Both the diluate and concentrate were fed into the ED stack at a flow rate of 3 L/min. An electrode rinse solution of 0.5 mol/L Na₂SO₄ was fed through the ED stack at a flow rate of 2.5 L/min, in order to prevent the generation of chlorine or hypochlorite. After each experiment, the diluate and concentrate stream were mixed and returned to the system as feed (diluate and concentrate) for the next experiment. EC was converted into total dissolved solids (TDS, mg/L) using a conversion factor $k = 0.64$ for Australian groundwater with high sodium content [25]. It is important to note however that this conversion is dependent on feed water characteristics.

Analytical Methods

Sampling was undertaken periodically from the recirculation line in the diluate and concentrate solution containers during each experiment. The pH, electrical conductivity and temperature of samples taken from the diluate and concentrate were measured using a pH/Conductivity meter (Multiline P4 epoxy gel combination pH electrode, automatic temperature compensation immersion probe, WTW). Anion analysis was undertaken using a DX-500 ion chromatograph (DIONEX, Sunnyvale, CA, USA). Separation was achieved using an IonPac AS4A-5C analytical column and a mobile phase composed of 1.7 mM sodium bicarbonate/1.8 mM sodium carbonate delivered at a flow rate of 2 mL/min. Post-column eluent suppression was achieved using an anion self-regenerating suppressor (ASRS-1) operated at a current of 50 mA. Detection was via an ED40 electrochemical detector set at an output range of 30 µS. Instrument control and data collection were accomplished using PeakNet software. Cation analysis was performed with Inductively-coupled Plasma Optical Emission Spectroscopy (ICP-OES) (Perkin Elmer Optima 5300 DV) and Inductively-coupled Plasma Atomic Mass Spectroscopy (ICP-MS) (Agilent 7500ce).

The stack resistance across the ED stack was calculated using Ohms law:

$$R = \frac{E}{I} \quad (\text{Equation 1})$$

Where, R is the resistance (Ohm), E is the electrical potential (Volt) and I is the electrical current (Amperes), from the voltage and current read from the DC power supply.

Ion concentration removal (%) from the diluate was calculated from using the following equation:

$$\frac{C_{Di}^0 - C_{Di}^{hr}}{C_{Di}^0} \times 100\% \quad (\text{Equation 2})$$

Where, C_{Di}^0 is the initial ion concentration in the diluate and C_{Di}^{hr} is the ion concentration in diluate in the time period T^{hr} (hr).

Flux (J_i) from the diluate into the concentrate was calculated using the following equation:

$$J_i = \frac{(C_{Ci}^{hr} - C_{Ci}^0)}{A_m T^{hr}} \quad (\text{Equation 3})$$

Where, C_{Ci}^{hr} is the concentration within the concentrate in the time period T^{hr} , C_{Ci}^0 is the initial concentration within the concentrate and A_m is the membrane area used in transport.

A mass balance of all contaminants within the ED system was performed, using the following equation, to determine the mass deposit on the ion-exchange membranes:

$$m_F = m_D + m_C + m_{ads/dep} \quad (\text{Equation 4})$$

Where, m_F is the feed mass, m_D is the diluate mass, m_C is the concentrate mass and $m_{ads/dep}$ is the adsorbed/deposited mass.

Results and Discussion

Electrodialytic parameters and ED performance

The resistance across the ED stack as a function of time and voltage is shown in Figure 2A. The stack resistance is relatively stable in the beginning of the experiment followed by a significant increase towards the final stages of the experiment. This is especially noticeable in the experiment with an applied voltage of 18V. The gradual increase occurs as a result of the depletion of ions within the diluate solution circulating through the ED stack and the resultant lowering of the current (amperes). The final stack resistance achieved in experiments with an applied voltage of 18V is higher than that achieved in experiments with an applied voltage of 12V, as a result of deeper demineralization of the diluate stream in the 18V experiment. This is illustrated in Figure 2B, which shows the concentration of total dissolved solids (TDS) within the diluate and concentrate. TDS removal in the 12V experiment was 80.5% compared with 94.9% in the 18V experiment, showing that higher applied results in increased TDS removal. The ADWG value of 0.500 g/L TDS was reached in the 18V experiment. This guideline was also achieved in the 12V experiment after the length of experimental time was extended to 110 minutes. The rapid increase in stack resistance in the 18V experiment can also be attributed to fouling of the ion-exchange membranes. In ED, fouling is the accumulation of contaminants, particulate, and organic matter that adhere to the membrane surface and/or within the pores of the ion-exchange membranes through hydraulic or other physical or electrical forces. As previously mentioned, fouling manifests itself as an increase in the electrical resistance, a decrease in the selectivity of the ion-exchange membrane and a decline in the flux of ions [20].

The mass of TDS (g) deposited on the membranes in the ED system is illustrated in Figure 2C. At each time period, the TDS mass deposited is greater in the 18V experiment. While some of this loss can be attributed to transport into the electrode rinse, the majority of this would be due to deposition on the ion-exchange membranes. As can be seen the amount of TDS deposited on the membrane was higher in the 18V experiments (6.9g) than in the 12V experiment (5.8g). The negatively-charged trace contaminants within the diluate solution, for example fluoride, would be deposited on the anion-exchange membranes and the positively-charged contaminants, for example calcium, deposited on the cation-exchange membranes. This deposition would also be dependent on other conditions such as trace contaminant solubility and solution pH.

Figure 2D shows the flux of TDS ($\text{g/m}^2/\text{h}$) from the diluate into the concentrate as a function of time. TDS flux ($\text{g/m}^2/\text{h}$) was greater in the 18V experiment in comparison to the TDS flux in the 12V experiment. After 40 min the TDS flux in the 18V experiment decreases steeply. This is a result of the deposition of contaminants on the ion-exchange membranes and the subsequent reduction in the transport of ions through the membranes. Further results on the flux of individual trace contaminants will be discussed later.

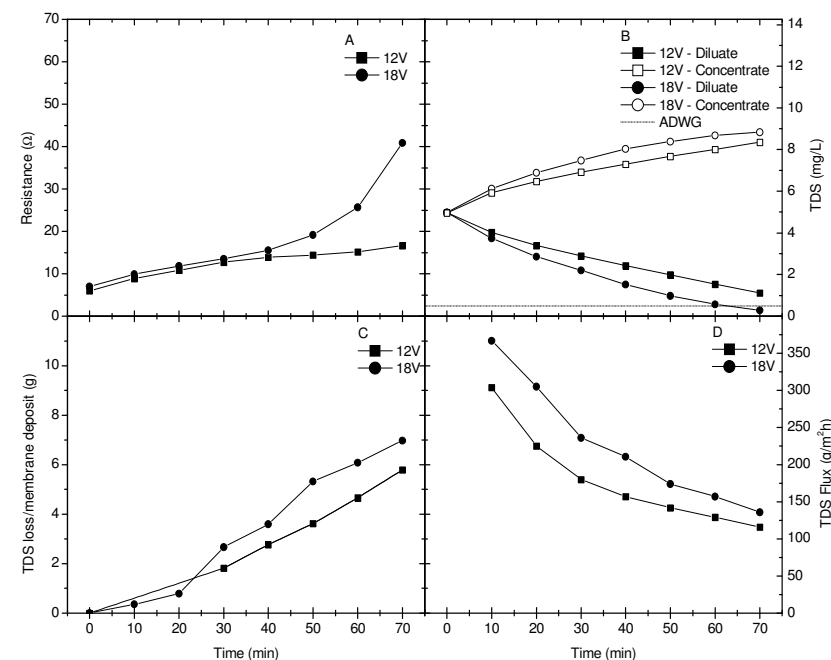


Figure 2. (A) ED stack resistance, (B) TDS (g/L), (C) TDS loss/membrane deposit (g), and (D) TDS flux ($\text{g/m}^2/\text{h}$) as a function of time and applied voltage. Also plotted is the ADWG (B bottom: 0.5 g/L) [4]

Trace contaminant removal

The removal of trace contaminants (anions and cations) from the Pine Hill bore water is shown in Table 2 and Figure 3. Bromide was removed to below detection limits in the 12V experiment (Table 2, Figure 3A) compared to 55.8% in the 18V experiment. A similar trend was also noted in regards to the other anions studied. The percentage removal of chloride (95.8%), fluoride (62.1-88.0%) and nitrate (80.6-81.5%) achieved in the 12V experiment (Table 2, Figure 3A) was also greater than in the 18V experiment (Table 2, Figure 3B). The transport of ions in ED is influenced by the concentration of the competitive ions within solution, whereby transport is increased if the concentration of competitive ions within solution is reduced. While the final TDS concentration in the 12V experiment was 1.11 g/L at 70 min, the concentration in the 18V experiment was 0.29 g/L. Therefore, theoretically the transport of ions in the 18V experiment should have been enhanced in comparison with the 12V experiment. Previous studies have showed increased removal with increased potential [28, 29]. This study however was undertaken on real bore water and no fouling prevention protocols were used. The amount of TDS deposited on the ion-exchange membranes during the 18V experiment was significantly higher, possibly hampering the transport of the anions through the anion-exchange membranes and into the concentrate. Results on the mass of trace contaminants deposited on the ion-exchange membranes and the flux of the trace contaminants through the ion-exchange membranes is outlined in the following section. The percentage removal of sulphate was relatively similar in both experiments (12V: 52.7%, 18V: 55.3%).

The removal of cations from the Pine Hill bore water is shown in Figure 3C (12V) and Figure 3D (18V). These results showed similar trends in removal as with the anions. Between 92.1-98% and 94.2-99% removal for magnesium and strontium, respectively, was achieved in the 12V experiment compared with 66.9% and 81.5% in the 18V experiment. The ED process was successful in the removal of strontium as a result of the relatively low initial concentration in the bore water (2.281 mg/L). Complete removal of strontium in the 12V experiment was reached after 80 min. The removal of calcium and potassium was also lower in the 18V experiment than in the 12V experiment.

Table 2. Removal of trace contaminants from the diluate as a function of applied voltage. DL refers to the analysis detection limit

| TRACE CONTAMINANT | REMOVAL (%) | |
|----------------------|-------------|------|
| | 12V | 18V |
| Bromide | 100 (< DL) | 55.8 |
| Calcium | 90.6-95.9 | 75.6 |
| Chloride | 95.8 | 67.6 |
| Fluoride | 62.1-88.0 | 68.1 |
| Magnesium | 92.1-98.0 | 66.9 |
| Nitrate | 80.6-81.5 | 62.3 |
| Potassium | 76.4-84.7 | 63.3 |
| Strontium | 94.2-99.9 | 81.5 |
| Sulphate | 52.7 | 55.3 |

Results from this study showed that ED was successful in removing chloride, fluoride and sulphate to concentrations below the ADWG values [24]. The final concentration of chloride and sulphate in the diluate were 96.5 mg/L and 392.3 mg/L, respectively, compared to the aesthetic guideline value of 250 mg/L and 500 mg/L. The final concentration of sulphate in the 18V experiment (365.2 mg/L) was also below the guideline value. While no guideline values have been reported for calcium and potassium, the final concentrations in the diluate in the 12V experiment were greatly reduced from their initial concentration in the bore water (Table 1) with 4.3-9.7 mg/L for calcium and 3.7-5.6 mg/L for potassium. In the 18V experiment the final concentrations in the diluate were 25.2 mg/L and 20.2 mg/L, respectively.

Overall, Figure 3 shows different kinetics for the anions and cations, with the removal of cations being significantly greater in the beginning stages of the ED process compared with a more steady increase in the removal of anions as a function of time. This difference in kinetics and removal trends may be the result of different ionic characteristics which are under current investigation.

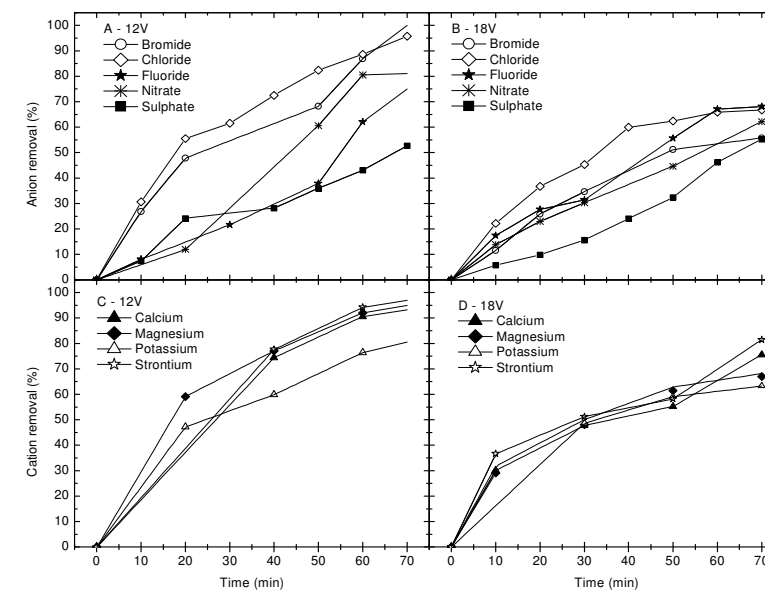


Figure 3. Removal of trace contaminants from the Pine Hill Bore water as a function of time and voltage. (A) Anion removal 12V, (B) Anion removal 18V, (C) Cation removal 12V, and (D) Cation removal 18V.

Trace contaminant loss/membrane deposition

Figure 4 shows that the mass of trace contaminants deposited on the ion-exchange membranes increases as a function of time. Understandably, the mass of ions deposited on the membranes is directly related to both the initial concentration within the bore water and also the applied voltage. The mass of ions deposited on the membranes was higher in the 18V experiment than in the 12V experiment due to the higher initial flux of ions through the ion-exchange membranes. The mass of potassium deposited on the membrane in the 18V experiment (90.5 mg) was considerably higher than in the 12V experiment (7.1-8.5 mg). The mass of chloride (12V: 1868 mg, 18V: 4339 mg) and sulphate (12V: 679 mg, 18V: 878 mg) deposited on the membrane is considerably greater than the other contaminants due to the high initial concentration within the Pine Hill bore water. This is also the case for calcium (12V: 332-514 mg, 18V 634 mg) and magnesium (12V: 294-375 mg, 18V: 406 mg) ions. This membrane deposit would form a fouling layer (scaling) at the membrane surface. As previously mentioned, scaling (fouling) manifests itself as an increase in the electrical resistance, a decrease in the selectivity of the ion-exchange membrane and a decline in the flux of ions [20]; which was demonstrated in the results from this study.

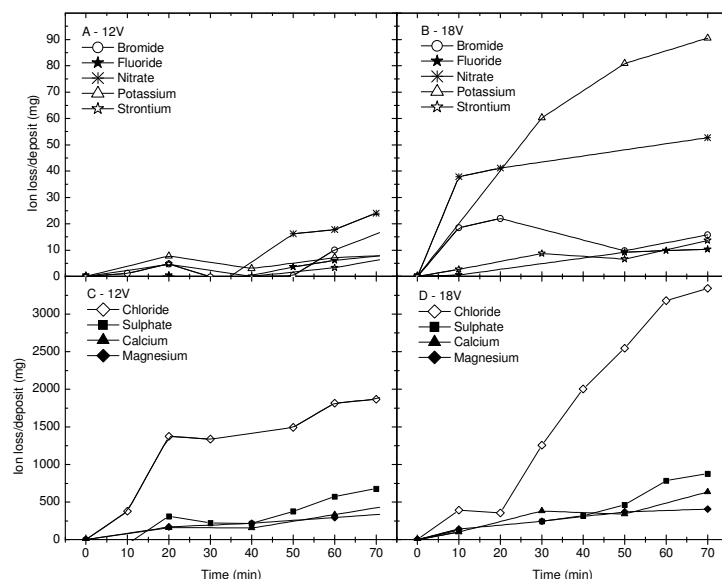


Figure 4. Ion loss/membrane deposit as a function of time and applied voltage. (A) Anion deposit 12V, (B) Anion deposit 18V, (C) Cation deposit 12V, and (D) Cation deposit 18V.

Conclusions

The results from this study demonstrated the performance of an electrodialysis system in the treatment of groundwater of high salinity and trace contaminant concentration from an isolated community in Central Australia. The removal of total dissolved solids was successful in that the treated bore water was below drinking water quality guideline levels. The applied voltage had an influence on the removal of the trace contaminants from the bore water, with 12V overall being the most effective. Results also demonstrated that electrodialysis was effective in reducing the concentration of the contaminants chloride, fluoride, sodium and sulphate to below guideline levels. However, fouling of the ion-exchange membranes is a critical issue. The deposition of trace contaminants on the ion-exchange membranes had an influence on the overall stack resistance throughout the experiments with a significant increase occurring in the later stages of the electrodialysis process. This increase in stack resistance was due to the formation of a fouling (scaling) layer on or in the membranes. This fouling layer in turn influenced the flux of total dissolved solids and trace contaminants from the diluate into the concentrate. This has an influence on the practical applicability of electrodialysis in the treatment of waters from remote communities in developing regions and further work is required to control such fouling.

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